

### **REMARKS**

Applicants wish to thank Examiners Peter J. Lish and Stuart Hendrickson for the very helpful and cordial interview on Tuesday, August 24, 2004. The following remarks include the gist of what was discussed at the interview.

At the outset, it should be noted that applicants' invention is directed to an activated carbon, suitable for use in electric double layer capacitors, which activated carbon is necessarily produced by carbonization of coconut shell and is activated by processes which cause the final activated carbon to have certain physical and electrical characteristics, which physical and electrical characteristics are not described in the prior art. Steam activation is preferred and has been demonstrated in the specification.

Applicants' claims are directed to a selection type invention wherein an activated carbon having specific surface area, pore diameter and pore volume of pores having diameters of 50 Å to 300 Å is produced for a specific application, i.e., electric double-layer capacitors having large output density per unit volume even when subjected to repeated charge-and-discharge cycles.

Applicants' claims require the activated carbon to have a very high BET specific surface area of from 2000 m<sup>2</sup>/g to 2500 m<sup>2</sup>/g. In addition, the material has a specific average pore diameter of from 1.95 nm to 2.20 nm (19.5 Å to 22.0 Å). This pore diameter is on the border of the micropore/mesopore interface. Further, the product contains some mesopores (50 Å to 300 Å) which have a specified pore volume. Further, the pore volume of pores having a pore diameter calculated according to the Cranston-Inkley method of 5.0 nm (50 Å) to 30.0 nm (300 Å) is from 0.05 cm<sup>3</sup>/g to 0.15 cm<sup>3</sup>/g. Applicants' claims also require the product to have a specific rest potential versus a lithium electrode in a non-aqueous electrolytic solution.

Applicants have achieved the superior activated carbon using a preferred process which involves (a) carbonization of a **specific starting material**, i.e., coconut shell, (b) activating the carbonized product at specified temperatures, preferably using **steam activation**, and (c) ensuring that the average pore diameter is quite small, i.e., 1.95 nm to 2.20 nm but also contains mesopores.

**The Rejection under 35 U.S.C. § 112.**

Claims 13-15 stand rejected under 35 U.S.C. § 112, second paragraph.

Claims 13-15 have been amended to obviate this rejection. As currently drafted, the pore volume set forth in claim 13 is of a narrower range ( $0.07 \text{ cm}^3/\text{g}$  to  $0.13 \text{ cm}^3/\text{g}$ ) than that set forth in claim 1 ( $0.05 \text{ cm}^3/\text{g}$  to  $0.15 \text{ cm}^3/\text{g}$ ). The pore volume of pores in both claim 1 and claim 13 is the volume of those pores of the carbonized material which have a pore diameter of 5.0 nm to 30.0 nm.

Because the basis for measuring pore volume in both claims 1 and 13 is the same, the pore volume range of claim 13 is clearly narrower than that of claim 1.

**The Rejection over Baker et al., U.S. Patent No. 5,416,056 (Baker '056)**

Of the references cited by the Examiner, the Baker '056 reference is the most informative regarding the direct relationship between the pore structure of carbonized carbon and the pore structure of the carbonized product. Simply stated, Baker teaches that, (a) ordinarily, activated carbons maintain the original macro structure of the starting carbonaceous material (col. 1, lines 55-61), and (b) the activation process affects the pore size of the product (col. 2, lines 1-7 and 38-55). Claims 1, 2 and 4-21 stand rejected under 35 U.S.C. §102(b) as anticipated by or, in the alternative, under 35 U.S.C. §103(a) as obvious over Baker et al., U.S. Patent No. 5,416,056.

At the outset, it should be noted that the microporous activated carbon product of Baker et al. are not designed for electric double layer capacitors. Baker '056 is concerned with producing an activated carbon product which is essentially microporous in structure such that and is characterized by a high storage capacity. The activated carbon product of Baker is characterized by greater than 80% of the total pore volume comprising pores of less than 20 Å in width and greater than 95% of the total pore volume comprising pores of less than 50 Å in width.

Because Baker '056 is concerned with methane absorption and the storage of methane in the activated carbon products, Baker is concerned with obtaining very high pore volume.

Baker suggests using wood as the lignocellulose material which is carbonized and activated. Baker recognizes that other materials may be used to make activated carbon. Included among the other materials is "nut shell (like almond shell and coconut shell)." (It should be noted that Baker appears to evidence that one skilled in the art would consider nut shell lignocellulosic materials as encompassing both almond shell and coconut shell. See col. 1, lines 45-61 of Baker '056 which is part of the "description of the prior art" section of the patent specification.)

Importantly, Baker advises that:

None of these activated carbons or other known carbons, however, achieved the desired objective of providing 150 v/v of deliverable gas capacity of 500 psi. At this storage pressure, known commercial carbons have achieved, at best, about 120 v/v of deliverable gas capacity. Therefore, the objective of this invention is to provide a highly microporous activated carbon of improved capacity (i.e., greater than 120 v/v at 500 psi) and preferably, a further objective is that the carbon is capable of meeting the industry target (i.e., 150 v/v) for deliverable capacity of natural gas stored on activated carbon at 500 psi.

Baker achieves his objective, not by activating nut shell material but by carbonizing and activating sawdust. Baker carbonizes the material and activates using a unique combination of phosphorous acid and potassium hydroxide. Neither of which is satisfactory alone. See col. 3,

lines 33-60. Accordingly, Baker's invention is a "two-stage activation product characterized by a pore volume of greater than 0.7 cc/g in pores of less than 20 Å in width. (Col. 4, lines 10-14).

It is respectfully submitted that the pore volume of Baker's pores is considerably greater than the pore volume set forth in applicants' claims. Applicants' claims define the pore volume of pores having a pore diameter of 50 Å to 300 Å as 0.05 cc/g to 0.15 cc/g, whereas the Baker material has a minimum pore volume of 0.7 cc/g in pores of less than 20 Å in width. This is a significant difference in pore volume.

The Examiner's attention is directed to Tables 1 and 2 of the Baker '056 reference which appear at cols. 5 and 6 respectively. Table 1 points out that the pores which are less than 1000 Å in width have pore volume ranging from 0.51 cc/g (TSA-17, Table 2) to 1.34 cc/g (TSA-22, Table 2). These pore volumes are dramatically higher than the pore volumes set forth in applicants' claims.

Several of Baker's examples describe activated carbon outside applicants' claims from the viewpoint of surface area. For example, TSA-17 and TSA-31 in Table 2 and TSA-16 and TSA-15 in Table 3, all have surface areas below the range set forth in applicants' claims.

It should be noted that throughout the Baker specification, the carbonaceous material utilized to prepare the products is described as either "sawdust" (col. 4, line 29) or "wood-base" (col. 4, line 22, col. 6, line 35 and col. 8, line 35). Baker teaches that the starting material used to make an activated carbon is highly material. Baker teaches that, ordinarily, activated carbons maintain the original macro structure of the starting material (col. 1, line 55 through 61). It is respectfully submitted that it would not have been obvious to one of ordinary skill in the art to utilize a nut shell material as a substitute for the wood material of Baker because there is no

suggestion in the Baker reference to do so. This is especially so when one considers that the pore structure is said by Baker to be that obtained from the basic material.

Additionally, applicants' claims define a product achieved via a steam activation process. Baker clearly teaches that the activation process affects the pore size of the product. See for example, col. 2, lines 1-7 and the discussion at col. 2, lines 38-55 which indicate that none of the activation processes of the prior art achieve the desired objective of producing 150 v/v of deliverable gas capacity of 500 psi.

Baker clearly teaches that activation of the carbonaceous precursors with phosphoric acid alone creates wide pores that not useful for methane absorption. Baker relied on activation using a combination of phosphoric acid and potassium hydroxide to produce both carbonization and activation in the same heating process. Next, after removal of acid from the system, the product was chemically activated further by blending with potassium hydroxide. See col. 3, lines 50-55. This is a very complex activation process and there is no suggestion in Baker to change the process. Indeed, as set forth by the court in *In re Freed*,

. . . it seems more logical and reasonable to infer that one teaching a chemical reaction process would set out the *least* number of reactions thought necessary to accomplish the desired objective. Thus, one skilled in the art who reads the teaching would have to presume that the reactants were not combined in the manner shown, some adverse side reaction or if no reaction would occur. *In re Freed*, 165 USPQ 570 (CCPA 197).

It is respectfully submitted that it is logical to assume that to one of ordinary skill in the art the teaching of Baker would not substitute sawdust for the wood used therein and, importantly, would not have used a steam activation process to achieve a significantly different product. Rather, one would assume that one needed to use Baker's raw material and Baker's unique activation process to obtain Baker's desired production.

Applicants' claims are directed toward a material suitable for a completely different purpose than that of Baker's material. Thus, there is no incentive to modify Baker's material to suit applicants' purpose.

**The Rejection over Adachi et al., U.S. Patent No. 5,430,606 ("Adachi '606").**

Claims 1-2 and 4-22 stand rejected under 35 USC § 102(b) as anticipated by, or, in the alternative, under 35 USC § 103(a) as obvious over Adachi et al., U.S. Patent No 5,430,606.

It is respectfully submitted that the Adachi '606 patent does not include teachings which either anticipate or make obvious the claimed invention. Indeed, on close analysis, the Adachi reference teaches against the claimed invention.

The Examiner is correct in indicating that Adachi '606 teaches an activated carbon for use in electric double layer capacitors, which activated carbon is made from coconut shells as the raw material. Additionally, Adachi teaches that by varying the temperature of heat treatment, one can obtain a BET surface area of 2001m<sup>2</sup>/g or 2337 m<sup>2</sup>/g, as set forth in the Examiner's claims. However, as recognized by the Examiner, Adachi has no specific suggestion regarding the pore diameter and pore volume of the activated, carbonized coconut shells.

Importantly, Adachi preferred and used a chemical activation process whereas applicants preferred and used a gaseous, steam activation process. On careful review, the Adachi '606 patent teachings indicate that the BET surface area of most activated carbons prepared therein is considerably less than 2000 m<sup>2</sup>/g. (See the Examples.) Additionally, Table 2 at col. 4 of Adachi evidences that the BET surface area in m<sup>2</sup>/g is controlled by the heat treatment and temperature of the chemical activation process use therein.

Adachi required activation heat treatments of above 700° C to obtain the high BET surface area that falls within the scope of applicants' claimed product. Unfortunately, however,

activation heat treatment at temperatures of about 700° C caused a significant drop in the capacitance of the product and resulted in an inferior, undesired product. (See col. 2, line 59 to col. 3, line 10). Because one of the objects of Adachi's invention is to obtain a high capacitance product, the Adachi reference strongly suggests accomplishing the heat treatment **"at a temperature below 700° C"** (col. 1, line 50 and line 54; see also col. 2, lines 59-68). Indeed, Adachi limited claim 1, the only claim, to a product obtained by heat treating at a temperature of 400° to 500° C.

The only carbonized coconut shell products in the examples of the Adachi '606 patent, which have the requisite surface area are runs 2-8 through 2-10 of Table 2, all of which have heat treatment temperatures in excess of 700° C. **These runs have poor capacitance and are said to be undesirable.** Adachi provides no data on the pore volume of these products.

Another aspect of applicants' invention which differs from that of Adachi '606 is the fact that applicants' prefer and exemplify a **gas activation** process wherein the carbonized coconut shells are **activated using steam**, preferably in an inert gas at temperatures in the range of from 800° C to 1300° C. This is an activation heat treatment not described and not suggested by the Adachi '606 patent. Indeed, Adachi '606 would teach activation heating at temperatures considerably less than 800° C.

It should be noted also that there is no teaching in the Adachi '606 patent of the pore volume limitation set forth in applicants' specification. Moreover, there is nothing in the Adachi '606 patent which would advise one of ordinary skill in the art that using the techniques described therein, one would achieve the desired pore volume. Indeed, the suggestion of Adachi is clearly to use a chemical (not steam) activating system and, importantly, to employ lower activation temperatures which yields products having a BET surface area considerably less than 2000 m<sup>2</sup>/g. See Runs 2-1 through 2-7 of Table 2.

It is respectfully submitted that the rejection over Adachi '606 is untenable and should be withdrawn.

**The Rejection Over Alford, U.S. Patent No. 5,926,361 ("Alford '361").**

Claims 1, 2 and 4-22 stand rejected under 35 USC § 102(e) as anticipated by or in the alternative, under 35 USC § 103(a) as obvious over Alford, U.S. Patent No. 5,926,361,.

Alford '361 is directed to a high power density double layer energy storage device. However, Alford '361 utilizes an activated carbon which is somewhat different from that claimed by applicants.

Alford does not specifically teach the use of coconut shell as a source of the carbon for activation. Alford's teaching of suitable raw material sources may be found at col. 3, lines 37-43 (which describes a number of materials including "nut shells", "nut pits" and the like. The Examiner has not provided evidence that the nut shells described by Alford are coconut shells. Accordingly, the Alford reference cannot be an anticipatory reference.

Importantly, however, "nut shells" are not the preferred material suggested and used by Alford as a precursor. Alford specifically states that "the more preferred precursors . . . [are] wood-based materials such as wood chips, wood flour, wood dust, sawdust and combinations thereof." (Col. 3, lines 40-43). Example 1 used sawdust. Example 2 also used sawdust. Hence, the Alford '361 reference does not and cannot anticipate. Importantly, Alford teaches away from the use of coconut shells as a source of carbon.

Assuming, *arguendo*, one of ordinary skill in the art would believe that the Alford teachings regarding "nut shells" would encompass coconut shell as well as other nut shells, Alford still does not anticipate or suggest the claimed invention. It is important to Alford that the lignocellulose based carbon have a pore volume of "greater than about 0.45 cc/g, a median pore width of 2.0 to 50.0 nm, a specific surface area of greater than about 500 m<sup>2</sup>/g, a mesopore



content, based on the total particle volume of greater than about 75% and a total combined micropore and macropore content based on the total particle volume of less than about 25%.

Pore sizes are defined in the Alford '361 specification at col. 1, lines 25-35, as follows:

Pore sizes are defined herein as **micropores** (pore width < 2.0 nanometers or nm), **mesopores** (pore width = 2.0 to 50.0 nm), and **macropores** (pore width > 50.0 nm). (Emphasis added).

According to applicants' claims, the average pore diameter is in the range 1.95 nm to 2.20 nm, such pores border on micropores. Alford, on the other hand, wishes to obtain a product wherein the median pore width is in the mesopore range. Indeed, the mesopore content based on the total particle volume of activated carbon is greater than about 75%.

Additionally, it should be noted that the Alford '361 carbon material is made by a significantly different process. Example 1 of Alford uses sawdust as a starting material. Applicants' process uses coconut shells. Additionally, Example 1 of Alford uses concentrated phosphoric acid during the carbonization heat treatment. This is a chemical activation process. Applicants' process (Example 1, page 21) does not activate using phosphoric acid but, rather, activates using an inert gas containing steam. Indeed, applicants' Example shows that when materials which were not coconut shell were treated to the same gaseous, steam activation concentration, the resultant product is significantly different. Additionally, applicants' product contains a significant and specified amount of oxygen in the activated product. There is no suggestion in the Alford patent that the final material should contain an active amount of oxygen.

Importantly, the Alford '361 patent process activates lignocellulose using a "chemical activation agent in a manner to effectively minimize the macropore and micropore structure of the activated carbon while maximizing the mesopore structure of the carbon." (Col. 3, lines 25-28) To this effect, the Alford patent describes the use of . . . a final densification step . . .

wherein the activated carbon is heated under an inert atmosphere and a temperature from about 900° C to about 1120° C. This densification step produces carbons with improved characteristics which make these carbons more suitable for use in CDLCs and previously known lignocellulosic materials (particularly wood-based lignocellulosic materials). (Col. 2, lines 22-34).

The heating step performed by Alford follows the chemical activation step and is described at col. 4, lines 12-17, as being a densification step wherein heating is conducted at a temperature from about 900° C to about 1120° C; with the preferred temperature range being from about 990° C to about 1120° C. This densification step, which follows the activation step, is a necessary aspect of the process provided by Alford and, importantly, affects the product. Indeed, it should be noted that applicants' specification ties in the bulk density to pore diameter. On page 10 of the specification, applicants state:

On the other hand, in case where the average pore diameter is too large, there results a decrease in bulk density of the activated carbon and a decrease in output per unit volume, thus such pore diameter not being preferred. The pore is preferably 2.0 nm (20.0 Å) to 2.15 nm (21.5 Å) . . . .

It should be noted that applicants' pores appear to be somewhat smaller than the mesopores desired by Alford.

Succinctly put, Alford uses a combination of chemical activation and subsequent high heat densification in order to make a product having a pore diameter significantly larger than that of applicants. All of applicants' claims contain the pore diameter limitation. Hence, it is respectfully submitted that Alford does not anticipate and does not make obvious the claimed invention.

**Rejection over Wennerberg, U.S. Patent No. 3,624,004 (“Wennerberg ‘004”)**

Claims 1, 2 and 4-21 stand rejected under 35 U.S.C. § 102(b) as anticipated by or, in the alternative, under 35 U.S.C. § 103(a) as obvious over Wennerberg, U.S. Patent No. 3,624,004 .

It is respectfully submitted that the Wennerberg reference is not directed to the use of activated carbon in an electrical capacitor. Further, the Wennerberg process teaches a process which forms the activated carbon by pyrolyzing petroleum coke acids and activating the carbonaceous material by gaseous activation with CO<sub>2</sub>. Wennerberg does not teach steam activation and the use of coconut shells as a starting material.

The Wennerberg reference is directed towards the production of fine surface area activated carbon with pore diameters that are significantly higher than the pore diameters of the activated carbon set forth in applicants' claims. Applicants' claims require the average pore diameter to be from 19.5 Å to 22 Å. The Wennerberg reference teaches the expansion of active carbons of 10 Å or less to pore diameters in the range of 20 Å – 100 Å. See col. 1, lines 30-36. The carbons are said to be “suitable to water purification applications and other possible separations of components in the solution.”

It is respectfully submitted that the activation process of Wennerberg ‘004 does not necessarily produce activated carbon of the requisite surface area. It should be noted that Examples III and IV indicate that a surface area in excess of 2000 m<sup>2</sup>/g is achieved only after activation with CO<sub>2</sub> for 60 to 90 minutes.

It should be further noted that Fig. 2, which is discussed by the Examiner does not lead one to the expectation that the activated carbon of Wennerberg exhibits the properties set forth in applicants' claims. Indeed, the activated carbon of Wennerberg has pore diameters within the range 20 to about 300 Å. Applicants' average pore diameter is only 19.5 to 22 Å.

Moreover, the porosity of the Wennerberg carbon is indicated at Fig. 2 to be 0.24 cc/g whereas that of applicants is considerably less, i.e., 0.05 cm<sup>3</sup>/g to 0.15 cm<sup>3</sup>/g for pores having a diameter of 50 Å to 300 Å. In other words, applicants' pore diameter appears to be more like the commercially available carbon than the carbon of the Wennerberg invention.

The Examiner has taken the position that the method of making the activated carbon does not limit the material itself. It is respectfully submitted that this is clearly not the case. The prior art, when taken as a whole, strongly indicates that the method of making the carbon and the raw material used to prepare the carbon and the method of activation of the carbon has a major affect on the ultimate properties of the carbon. On this point, see Baker '056, col. 1, lines 55 through 61.

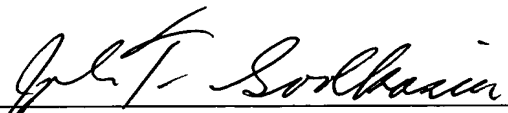
It is respectfully submitted that the claims of this application as amended are now in condition for allowance and early indication of allowability is requested.

Respectfully submitted,

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